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Environmental factors affecting methane distribution and bacterial methane oxidation in the German Bight (North Sea)

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ABSTRACT

River estuaries are responsible for high rates of methane emissions to the atmosphere. The complexity and diversity of estuaries require detailed investigation of methane sources and sinks, as well as of their spatial and seasonal variations. The Elbe river estuary and the adjacent North Sea were chosen as the study site for this survey, which was conducted from October 2010 to June 2012. Using gas chromatography and radiotracer techniques, we measured methane concentrations and methane oxidation (MOX) rates along a 60 km long transect from Cuxhaven to Helgoland. Methane distribution was influenced by input from the methane-rich mouth of the Elbe and gradual dilution by methane-depleted sea water. Methane concentrations near the coast were on average 30 ± 13 nmol L^{-1}, while in the open sea, they were 14 ± 6 nmol L^{-1}. Interestingly, the highest methane concentrations were repeatedly detected near Cuxhaven, not in the Elbe River freshwater end-member as previously reported. Though, we did not find clear seasonality we observed temporal methane variations, which depended on temperature and presumably on water discharge from the Elbe River. The highest MOX rates generally coincided with the highest methane concentrations, and varied from 2.6 ± 2.7 near the coast to 0.417 ± 0.529 nmol L^{-1} d^{-1} in the open sea. Turnover times varied from 3 to >1000 days. MOX rates were strongly affected by methane concentration, temperature and salinity. We ruled out the supposition that MOX is not an important methane sink in most of the Elbe estuary and adjacent German Bight.

1. Introduction

Methane is the most abundant organic gas in the atmosphere. Being a potent greenhouse gas, it plays an important role in warming the Earth's atmosphere (Kirschke et al., 2013). Its contribution to global warming is attenuated by comparatively low concentrations and short lifetime in the atmosphere. Methane has the second-largest radiative forcing of the long-lived greenhouse gases, after CO_2 (Ramaswamy et al., 2001). The atmospheric concentration of methane has increased to a level unprecedented in at least the last 800,000 years (Stocker et al., 2013). Its average concentration nowadays is around 1.8 ppm (Kirschke et al., 2013). Changes in methane concentration, which are defined by the balance between sources and sinks of methane, have led to investigations on the microbial-driven methane cycle in various environments.

About 60% of the methane released to the atmosphere is from anthropogenic sources such as agriculture, waste treatment, biomass burning, and fossil fuels. The remaining 40% originates from natural sources, mainly wetlands (Kirschke et al., 2013). Among these sources, the world's oceans contribute <0.1–10% of the methane emissions (Scranton and McShane, 1991; Bates et al., 1996; Middelburg et al., 2002; Conrad, 2009). Digestive tracts of zooplankton (Bianchi et al., 1992; De Angelis and Lee, 1994), and

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organic particulate matter (Karl and Tilbrook, 1994) are the main methane sources in the open ocean. However, about 75% of global oceanic methane emission occurs in shelf areas and estuaries (Bange et al., 1994; Bange, 2006). The main sources of methane in these areas are sediments, adjacent rivers, tidal flats, and marshes (Bange et al., 1994; Bange, 2006; Abril et al., 2007; Grunwald et al., 2009; Bussmann, 2013). Sedimentary release of methane which follows methanogenesis (Koch et al., 2009; Bussmann, 2013) and abiotic methane production (Hovland et al., 1993) is supplemented with lateral methane transport. Water discharge from rivers, as well as tidal influence, are factors that greatly control methane distribution (Rehder et al., 1998; Grunwald et al., 2009). Thus, these areas represent diverse and complex hydro-dynamic systems. Besides, estuarine systems are subject to significant seasonal changes (Sansone et al., 1998; Middelburg et al., 2002; Abril and Borges, 2005), a factor which is not considered in many studies. Bacterial methane oxidation, along with degasing and dilution of methane-rich water, are also important factors controlling methane distribution in the water column (Grunwald et al., 2009). Up to 80–90% of the available methane can be metabolized and thereby disposed of by bacterial methane oxidation in the freshwater (Reeburgh et al., 1993; Guérin and Abril, 2007). In the marine environment, however, methane oxidation (MOX) rates are in general lower (Valentine et al., 2001; Mau et al., 2013). Though, knowledge on bacterial methane oxidation in the water column rapidly improves, MOX rates measurements in estuaries are still scarce. Additionally, an improved knowledge of the environmental factors controlling bacterial methane oxidation and the physiological properties of these organisms is crucial and this topic needs further investigation (Valentine, 2011; Mau et al., 2013).

The objectives of this study were to describe the spatial and seasonal variations of methane in the German Bight, near the Elbe estuary. The methane concentration, as well as several hydrochemical parameters from the bottom and surface waters, were measured over a period of two years along a 60 km long transect in the German Bight. Along with temperature, the concentrations of inorganic nutrients (ammonium, nitrite, nitrate, phosphate, and silicate) and suspended particulate matter (SPM) were measured. Salinity was an especially important parameter, as it is a direct indicator of the extent of water discharge of the Elbe River. The aim of this study was to bring the importance of bacterial methane oxidation as a significant methane sink into focus. Therefore, we made highly sensitive radiotracer measurements to estimate MOX rates in the water column (Valentine et al., 2001). Only on the basis of these comprehensive examinations we were able to determine the main environmental factors that control methane distribution and MOX rates.

2. Materials and methods

2.1. Study area

The North Sea (including its estuaries and fjords) has a surface area of about 750,000 km^2 and a volume of about 94,000 km^3 (Commission, 2000). The non-tidal circulation of the North Sea is dominated by a cyclonic residual current. Water from the Atlantic (Fair Isle Current, Shetland Flow) flows southward along the British coast and returns northward, together with influxes from the English Channel and various rivers along the coasts of the Netherlands, Germany, and Denmark (Rehder et al., 1998). The German Bight is the south-eastern bight of the North Sea, bounded by the Netherlands and Germany to the south, and Denmark and Germany to the east (the Jutland peninsula). To the north and west, it is bounded by the Dogger Bank. The depths in this area vary mainly from 20 to 40 m (Czitrom et al., 1988). The German Bight consists mainly of a mixture of the Central (Southern) North Sea water masses and continental coastal waters (Becker et al., 1992). The water column in the central (southern) North Sea can be stratified into two slightly different layers (Czitrom et al., 1988; Becker et al., 1992). Inshore water did not show any stratification in either summer or winter. Freshwater discharge from the Elbe and Weser rivers causes a large salinity contrast near the shore (Czitrom et al., 1988). Analysis of horizontal density gradients did not show a clear annual cycle either near the shore or offshore (Czitrom et al., 1988). Surface sediments affected by tidal or residual current, wave action, and heavy bottom trawling are very mobile (ICES, 1988; Becker et al., 1992).

The Elbe is one of the major rivers of central Europe, with a total catchment area of about 150,000 km^2. It runs from the Czech Republic through Germany, and reaches the German Bight in its north-eastern region, near Cuxhaven. The Elbe's mean annual discharge at the river mouth is 860 m^3 s^{-1}. The discharge regime is mainly controlled by rainfall and snowmelt, therefore it peaks in April/May (Simon, 2005).

2.2. Water sampling

Samples were collected for 2 years from 2010 to 2012. Eleven one-day sampling cruises took place on the 7.10, 8.12 in 2010, 12.01, 2.03, 4.05, 6.07, 29.09 in 2011 and 11.01, 29.02, 28.03, 11.06 in 2012. All sampling procedures and some of the processing of the samples were done on board the research vessels 'Uthörn' and 'Ludwig Prandtl'. Seven sampling stations were distributed along the Helgoland–Cuxhaven transect (Fig. 1). The names of the stations were determined by their distance from the northernmost coastal point near Cuxhaven, so the most south-eastern station was denoted 'Sea kilometre (Sk) 1', and the most north-western, near Helgoland, 'Sk 59'. We determined stations Sk 1, Sk 14, and Sk 20 to be 'coastal stations', and stations Sk 27, Sk 32, Sk 49, and Sk 59 to be 'marine stations'. Water samples were collected with Niskin bottles from the water surface (1 m below the surface) and from the bottom (1 m above the bottom).

2.2.1. Oceanographic parameters

Water temperature was measured to monitor seasonality during the study period. Salinity was measured to account for the proportion of freshwater from the Elbe River in North Sea water. Oxygen concentration was measured to investigate its effect on MOX rate, and accordingly, on methane distribution. During the Prandtl cruises, temperature, salinity, and oxygen in the water column were measured immediately after sampling on board using a Universal Pocket Meter (Multi 340i) with precisions of 1% for salinity, 0.1 °C for temperature, and 0.5% for oxygen. Salinity was measured in μS cm^{-1}, and then converted according to the Practical Salinity Scale. For the Uthörn cruises, a sea-bird CTD sensor was mounted to the water sampling rosette. In July 2011, temperature measurements were not made due to technical problems. Thus, we obtained temperature data from the database of the River Basin Community Elbe (RBC Elbe; in German, 'Flussgebietsgemeinschaft (RBC) Elbe; http://www.fgg-elbe.de/elbe-datenportal.html). These data were collected near Cuxhaven two days before our cruise. Comparisons of RBC measurements with ours for other months did not reveal any significant difference (±1 °C). These data from July were excluded from the comparison of temperatures on the surface and on the bottom, but were used for the correlation analysis between temperature and other factors.

2.2.2. Suspended particulate matter (SPM)

Sampled water was filtered using pre-washed and pre-weighed GFC filters (Whatman TM), which were afterwards dried for 24